Dynamic response of 1.3-μm-wavelength InGaAs/GaAs quantum dots

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The temperature-dependent dynamic response of 1.3-μm-InGaAs/GaAs quantum dots is investigated using time-resolved photoluminescence upconversion with subpicosecond temporal resolution for excitation in either the GaAs bulk region surrounding the dots or within the wetting layer. Relaxation to the quantum-dot ground state occurs on a time scale as short as 1 ps, while radiative lifetimes as short as 400 ps are measured. The influence of nonradiative recombination is observed only for temperatures above 250 K. At temperatures below 77 K, an increase in the relaxation time and lifetime is observed when carriers are injected into the bulk GaAs region versus excitation into the wetting layer, which suggests that diffusion in the bulk GaAs region influences both the relaxation rate and the recombination rate.
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Quantum-dot (QD) active regions are rapidly advancing for use in lasers and microcavity light emitting diodes[1–8] because they can have much lower threshold current density,[6,8] wavelengths that extend beyond 1.3 μm,[5–8] and reduced temperature sensitivity as compared to planar quantum-well lasers. The QDs also provide the three-dimensional electronic confinement needed for microcavity and photonic band-gap lasers and light emitters. The modulation rate of a QD microcavity light emitting diode will be limited by its spontaneous lifetime, and under ideal conditions and low temperatures the microcavity can shorten the QD radiative lifetime by an order of magnitude and obtain high efficiency.[9–11] For lasers, the modulation rate might be limited by quantum capture and relaxation in the QD's discrete energy levels.[12] These processes can be slower than in a planar quantum well due to severe restrictions imposed on energy relaxation routes by the discrete nature of the QD energy levels. Measurements of the dynamic response can then be quite important to assess the potential of QD devices for high-speed modulation, and different types of QDs or QDs placed in different confining heterostructures may show different dynamic behavior.

A QD's size and potential depth, its wetting layer thickness and potential depth, and the dimensions of the bulk collection region surrounding the QD and its wetting layer all combine to set the electronic energy spectrum relevant to capture, relaxation, and spontaneous light emission. When the QD becomes large, the capture process must become identical to that for a planar quantum well. On the other hand, even for strong three-dimensional confinement, the QD capture and energy relaxation may resemble that in quantum wells, if the energy level spacing approaches the optical phonon energy. The QD's radiative lifetime also depends on size, especially if the emission is based on superradiance. For emission by superradiance, larger QDs are predicted to have shorter radiative lifetimes,[13,14] with the lifetime limited by electronic confinement in the shortest dimension. Previous studies show that InGaAs QDs with heights estimated to be 44 Å (room temperature ground state emission at ~1.13 μm) have minimum radiative lifetimes of ~1 ns.[15] Below, we show that similar 1.3 μm QDs with heights estimated to be ~150 Å exhibit minimum radiative lifetimes of ~400 ps.

Based on the criteria above, the 1.3-μm-InGaAs/GaAs QDs are attractive because of their large size, and they have the added benefit that their wavelength is important for fiber interconnects. So far, however, there have been no reports on the dynamic response of this type of QD. Studies on similar but shorter wavelength QDs were limited by the temporal resolution.[15]

The InGaAs/GaAs QDs are grown using a cycled submonolayer approach demonstrated for 1.3 μm lasers[5] and reported earlier for similar large QDs.[16–18] The QDs are formed from a deposition of 10 monolayers of $In_{0.5}Ga_{0.5}As$ average composition. They have a density of $\sim 10^{10}\,cm^{-2}$ and a lateral size of ~500 Å as measured by atomic force microscopy just after deposition, while cross-sectional transmission electron microscopy shows that the lateral sizes are ~300 Å after covering with GaAs. Two QD layers are grown within the AlGaAs/GaAs confinement heterostructure illustrated in the inset of Fig. 1. The QDs exhibit a single PL peak at low excitation and four well-defined peaks separated by 60–70 meV at high excitation.

Time-resolved photoluminescence (PL) measurements were performed using 140 fs pulses from a mode-locked Ti:sapphire laser tuned to either 809 or 840 nm. For 809 nm pumping, carriers are generated primarily within the 0.2-μm-thick GaAs region. In this case, the QD ground state emission is influenced by optically generated carriers that must diffuse or drift from the bulk GaAs regions into the wetting

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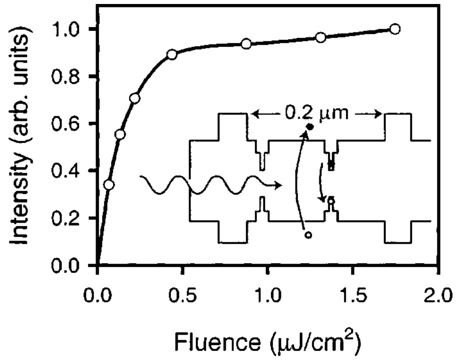

FIG. 1. Time-resolved ground state emission (measured 18 ps after excitation) vs pump level for $T=77$ K and 809 nm excitation. The peak ground state intensity saturates for peak pump fluences above ~0.5 μJ/cm^2. The inset shows a schematic illustration of the 1.3-μm-QD heterostructure.

layers before capture and relaxation to the ground state. For 840 nm pumping and temperatures below 150 K, carriers are excited primarily in the wetting layer and diffusion in the bulk GaAs region should play little if any role in the measured carrier dynamics.

The QD PL is upconverted in a 0.5-mm-thick $LiIO_3$ crystal with a system temporal resolution of approximately 200 fs and a 10 meV spectral resolution. All measurements were performed at the peak of the QD ground state emission, which varied from 1.21 to 1.31 μm over the range of 11 to 300 K. Figure 1 shows the 77 K ground-state PL intensity measured 18 ps after pumping at 809 nm. Ground-state filling is readily observed as indicated by the complete saturation of the PL for pump levels greater than 0.5 μJ/cm^2. Figure 2(a) shows the onset of the ground state PL intensity for a peak excitation fluence of 0.13 μJ/cm^2 and lattice temperatures varying from 77 to 250 K. The data are well characterized by a monoexponential rise time that increases from 1.5 ps at 77 K to 2.4 ps at 250 K. Such fast relaxation times suggest that carrier-carrier scattering plays a role in filling the ground state of these QDs.[19,20] However, our QDs are large enough so that the electron energy levels are separated by approximately an optical phonon energy (~40 meV), so that the relaxation rate may be increased due to optical phonon emission.

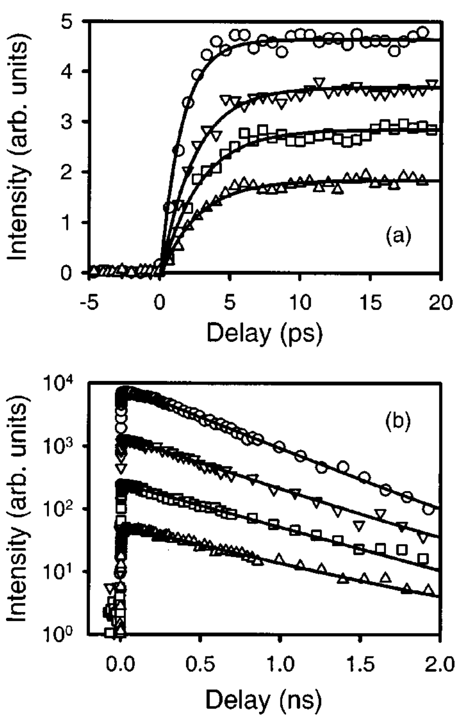

FIG. 2. (a) Ground state emission vs time on a picosecond time scale for 809 nm pumping and temperatures of 77 K (circle), 150 K (inverted triangle), 200 K (square), and 250 K (upright triangle) and for a peak pumping level of 0.13 μJ/cm^2. (b) Semilogarithmic plot of the ground state emission vs time on a nanosecond time scale for the same temperatures and excitation as in (a). Note that the amplitudes of both data sets have been adjusted for clarity. The solid lines are the results of monoexponential fits used to estimate the luminescence rise time (a) and decay times (b). The fits yield rise times of 1.5, 2.4, 2.7, and 2.4 ps and decay times of 440, 520, 580, and 700 ps at 77, 150, 200, and 250 K, respectively.

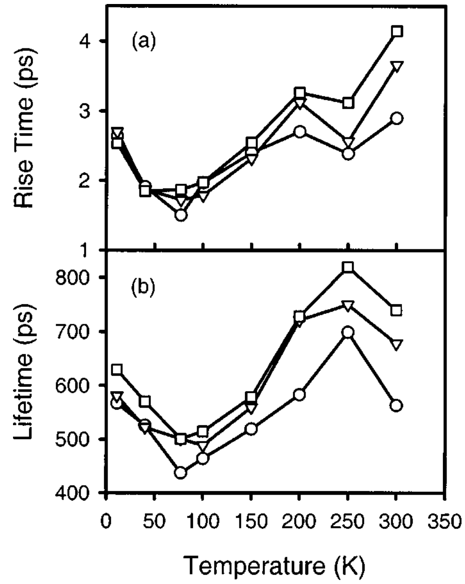

FIG. 3. (a) Ground state PL rise time vs temperature for 809 nm peak pump fluences of 0.13 (circle), 0.22 (inverted triangle), and 0.44 (square) μJ/cm^2. (b) Ground state PL decay time vs temperature for the same pump fluences as in (a).

Figure 2(b) shows the decay of the PL intensity. Lifetimes are extracted from decaying monoexponential fits to these data. In this range of temperatures, the carrier lifetime clearly increases with increasing temperature. The 77 K radiative spontaneous lifetime of 440 ps is shorter than that reported for InGaAs/GaAs[21–23] or InAs/GaAs QDs emitting at shorter wavelengths.[24–27]

The PL rise times measured with 809 nm excitation are plotted in Fig. 3(a) for several pump levels and a wider range of temperatures than shown in Fig. 2. The rise time is minimized in the 40–100 K range and then increases with increasing temperature. Initial modeling of the QD capture using the nonlinear rate equations in Ref. 28 indicates that, when there is a large disparity between the number of wetting layer states versus the number of levels in a QD, a temperature dependent rise time can occur due to thermal reexcitation of carriers from QD levels into the wetting layers.[28] Below 77 K, the increase in rise times is apparently due to diffusion-limited capture from the GaAs regions.

The decay times, shown in Fig. 3(b), indicate an increase in the radiative lifetime for temperatures from 100 to 250 K. Similar behavior has been observed in InGaAs/GaAs,[21] InAs/GaAs,[25–27] and InP/GaInP[29] QDs, and various explanations have been put forth. Within this temperature range we

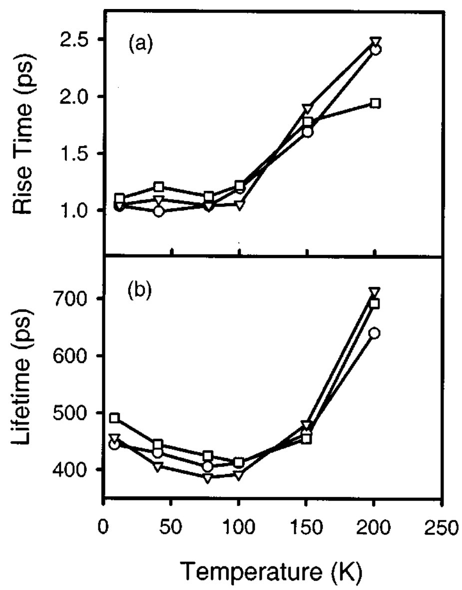

FIG. 4. (a) Ground state PL rise time vs temperature for 840 nm peak pump fluences of 0.87 (circle), 1.31 (inverted triangle), and 2.18 (square) $\mu J/cm^2$. (b) Ground state PL decay time vs temperature for the same pump fluences as in (a). For this pump wavelength, no significant saturation of the ground state PL is observed at these fluences.

observe significant emission from higher energy QD states even for excitation levels well below ground state saturation. Hence this effect may be explained by an increased occupation of higher energy levels with temperature, coupled with dipole transition selection rules. An observed decrease in lifetime above 250 K is associated with nonradiative recombination. The critical temperature for this effect is considerably higher than observed in shallower QDs.[21,26,27]

We also observe an increase in the decay times at the lowest temperatures for pump excitation into the bulk GaAs region. Again this might be explained by slow diffusion of carriers from the GaAs to the QD wetting layers. The separation between QD regions is $\sim 0.2\ \mu m$. The longest distance the carriers must diffuse is $\sim 0.2\ \mu m$, and if the diffusion time increases the decay time by ~ 200 ps the estimated diffusion coefficient is $\sim 2\ cm^2/s$. At 11 K the mobility estimated from the Einstein relation is on the order 2000 $cm^2/(V\,s)$, which seems reasonable for holes.

By using the 840 nm excitation for which carriers are generated primarily in the wetting layers, the low temperature diffusion effects can be nearly eliminated as shown in Fig. 4. For temperatures of 100 K and below, the rise time [Fig. 4(a)] is independent of temperature, and the lifetime [Fig. 4(b)] exhibits only a weak temperature dependence. We also observe that both the energy relaxation times and the carrier lifetimes tend to be shorter for 840 nm excitation, with measured minimum values of approximately 1 and 400 ps, respectively.

In summary, we present a systematic study of the dynamic response of 1.3-μm-InGaAs/GaAs QDs measured with subpicosecond resolution. Energy relaxation times as short as 1 ps and radiative lifetimes as short as 400 ps are observed. The short radiative lifetime for these large QDs is suggestive of superradiance, but further studies are required for a definitive conclusion. Nevertheless, the results illustrate that 1.3 μm QDs have potential for high-speed modulation for efficient microcavity light emitting diodes and lasers.

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